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(71) Applicant: Société Anonyme dite: NORSOLOR  
Tour Aurora, Place des Reflets  
F-92080 Paris la Défense 2, Cedex 5(FR)

(72) Inventor: Mitsuno, Tatsuyuki  
1-9, Yushidainishi  
Ichihara-shi Chiba(JP)  
Inventor: Shinonaga, Hideo  
285-172, Higashiterayama-cho  
Chiba-shi Chiba(JP)  
Inventor: Daimon, Akio  
1-9, Yushidainishi  
Ichihara-shi Chiba(JP)

(74) Representative: Dubost, Thierry  
c/o NORSOLOR Service Propriété  
Industrielle B.P. 57  
F-62670 Mazingarbe(FR)

(94) Filler-containing polypropylene resin composition and process for producing the same.

(57) The composition comprises (a) from 27 to 87 % by weight of a polypropylene resin, (b) from 3 to 15 % by weight of a modified polypropylene resin containing from 0.1 to 3 mol % of an unsaturated dicarboxylic acid or anhydride repeating unit, (c) from 5 to 30 % by weight of an ethylene copolymer composed of from 82 to 99.7 mol % of an ethylene repeating unit, from 0 to 15 mol % of an ester repeating unit selected from alkyl acrylates and alkyl methacrylates having from 1 to 8 carbon atoms in the alkyl moiety thereof, and from 0.3 to 3 mol % of an unsaturated dicarboxylic acid anhydride repeating unit, and (d) from 5 to 40 % by weight of a filler.

The molded products obtained by molding composition exhibit excellent finishing properties, such as adhesion, coating properties, and printability, as well as improved rigidity and thermal properties.

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# FILLER-CONTAINING POLYPROPYLENE RESIN COMPOSITION AND PROCESS FOR PRODUCING THE SAME.

This invention relates to a filler-containing polypropylene resin composition and a process for producing the same. More particularly, it relates to a filler-containing polypropylene resin composition having improved finishing properties and a process for producing the same.

In recent years, applications of filler-containing polypropylene resins to automobile parts and appliances have been markedly broadened because of their excellent mechanical and thermal properties.

In the application to automobile parts, for instance, interior parts, such as an instrument board, a trim, and a pillar-post, are often covered with non-woven fabric, foamed fabric, leather cloth made of polyvinyl chloride, and the like to produce an atmosphere of high-class. Further, there is noted a tendency to coat a bumper in color harmony with the body in a decorative style. In appliance parts, also, printing is commonly employed for decoration.

Polypropylene resins often find difficulty in finishing due to their poor adhesion or coating properties arising from non-polarity. It is well known to add fillers to polypropylene resins in order to improve these finishing properties. Incorporation of fillers improves finishing properties to some extent depending on the kind and amount of the filler, but such an improvement is still sufficient for practical use.

In order to further improve finishing properties, i.e., compatibility between polypropylene and adhesives or coatings, a medium called "primer", which typically comprises chlorinated polypropylene and toluene, etc., is usually provided therebetween. However, since the primer itself is expensive and an extra coating step is required, the cost of the final products is increased. Therefore, further improvements have been required.

In an attempt to improve surface properties, physical or chemical etching of the surface of polypropylene molded products prior to adhesion has been studied and put into practice. However, this process involves more complicated steps than the primer coating, and some conditions of etching cause deformation of molded products or produce insufficient effects.

In the case of coating, the process of coating a primer is being shifted to a process comprising oxidatively polarizing the surface of molded articles by irradiation with plasma to improve coating properties. Under the present situation, however, this process does not always achieve satisfactory results because the plasma irradiation should be effected in a batch system and the polarity obtained is non-uniform depending on the shape of the molded articles.

Attempts have also been made to impart polarity to polypropylene per se. For example, it has been proposed to modify polypropylene by grafting with an unsaturated carboxylic acid or anhydride, e.g. acrylic acid, maleic anhydride, etc., in the presence of a free-radical initiator (e.g. organic peroxides) or to blend the above-described modified polypropylene with unmodified polypropylene, as disclosed in Japanese Patent Publication No. 78149/75 and Japanese Patent Publication No. 10264/77. According to this process, however, grafting is accompanied by degradation of the polypropylene by the peroxide so that the amount of the unsaturated carboxylic acid or anhydride to be incorporated is limited to about 0.2 % by weight with respect to polypropylene. As a result, the surface modifying effect attained is insufficient. Whenever any effects are achieved, mechanical properties are impaired due to molecular weight reduction of polypropylene.

In order to overcome the disadvantages associated with the conventional polypropylene resins, there was also proposed a composition comprising a polypropylene resin, an ethylene copolymer comprising an ethylene unit, an ester unit selected from alkyl acrylates and alkyl methacrylates, and an unsaturated dicarboxylic acid anhydride unit, and a filler, as disclosed in European Patent Application No. 213,720. However the aforesaid filler-containing polypropylene resin composition shows insufficient improvement of rigidity and exhibits unsatisfactory thermal properties, although surface modifying effects can be achieved as expected.

One object of this invention is to provide a filler-containing polypropylene resin composition having excellent finishing properties, such as adhesion, coating properties, printability, and the like, as well as improved mechanical and thermal properties.

Another object of this invention is to provide a process for producing such a filler-containing polypropylene resin composition.

The inventors have conducted extensive investigations on improvements of the above-described filler-containing polypropylene resin composition of European Patent Application No. 213,720. As a result, it has now been found that marked improvements on rigidity and thermal properties can be obtained by incorporating a modified polypropylene resin containing an unsaturated dicarboxylic acid or anhydride

repeating unit into the aforesaid composition.

The filler-containing polypropylene resin composition according to the present invention comprises :

- (a) from 27 to 37 % by weight of a polypropylene resin,
- (b) from 3 to 15 % by weight of a modified polypropylene resin containing from 0.1 to 3 mol % of an unsaturated dicarboxylic acid or anhydride repeating unit,
- (c) from 5 to 30 % by weight of an ethylene copolymer comprising from 82 to 99.7 mol % of an ethylene repeating unit, from 0 to 15 mol % of an ester repeating unit selected from alkyl acrylates and alkyl methacrylates having from 1 to 8 carbon atoms in the alkyl moiety thereof, and from 0.3 to 3 mol % of an unsaturated dicarboxylic acid anhydride repeating unit, and
- (d) from 5 to 40 % by weight of a filler.

A preferred process for producing the above-described filler-containing polypropylene resin composition comprising components (a) to (d) comprises melt-mixing components (a), (b) and (d) to obtain (e) a mixture and then melt-mixing mixture (e) with component (c).

- The polypropylene resins which can be used in the present invention include crystalline polypropylene, crystalline propylene-ethylene block or random copolymers, crystalline propylene- $\alpha$ -olefin copolymers wherein the  $\alpha$ -olefin has from 4 to 10 carbon atoms, and mixtures thereof.

The polypropylene resin usually has a melt index (as determined according to ASTM D-1238) ranging from 0.5 to 100, and preferably from 1 to 20 g/10 min.

- The modified polypropylene resins which can be used as component (b) comprise the above-described polypropylene resin and from 0.1 to 3 mol % of an unsaturated dicarboxylic acid or anhydride repeating unit. Monomers providing the unsaturated dicarboxylic acid or anhydride repeating unit include maleic anhydride, citraconic anhydride, itaconic anhydride, tetrahydrophthalic anhydride, acrylic acid, methacrylic acid, etc., with maleic anhydride, acrylic acid and methacrylic acid being preferred.

- If the content of the unsaturated dicarboxylic acid or anhydride repeating unit in the component (b) is less than 0.1 mol %, effects of improving coating properties, printability, adhesion, and the like are unsufficiently produced. If it exceeds 3 mol %, the amount of polar groups introduced becomes excessive to deteriorate compatibility with the polypropylene resin.

- The modified polypropylene resin can be obtained by known techniques, such as a process comprising dissolving a polypropylene resin in an organic solvent and adding an unsaturated dicarboxylic acid or anhydride and at least one free-radical initiator thereto, followed by heating with stir ring, or a process in which all components are fed to an extruder to effect graft copolymerization.

- Monomers providing the ester repeating unit in the ethylene copolymer (c) of the present invention include methyl acrylate, ethyl acrylate, n-butyl acrylate, isobutyl acrylate, methyl methacrylate, ethyl methacrylate, 2-ethylhexyl methacrylate, butyl methacrylate, etc. (hereinafter referred to as unsaturated ester compounds). The ester repeating unit is present in the ethylene copolymer (c) in a proportion of from 0 to 15 mol %, and preferably from 3 to 12 mol %.

- Monomers providing the unsaturated dicarboxylic acid anhydride repeating unit of component (c) include maleic anhydride, citraconic anhydride, itaconic anhydride, tetrahydrophthalic anhydride, and the like, with maleic anhydride being particularly preferred. The proportion of the unsaturated dicarboxylic acid anhydride repeating unit in the ethylene copolymer (c) ranges from 0.3 to 3 mol %, and preferably from 1 to 3 mol %. With proportions lower than 0.3 mol %, the amount of polar groups which contribute to finishing is too small to produce satisfactory improving effects.

- Processes for producing the ethylene copolymer (c) are not particularly restricted and include known processes, such as high-pressure bulk polymerization, in which ethylene is copolymerized in the presence of at least one free-radical initiator at a temperature of from 100 to 300 °C under a pressure of from 500 to 3000 atm. in the presence or absence of an appropriate solvent or a chain transfer agent. The high-pressure polymerization may be suitably carried out by dissolving the above-described unsaturated dicarboxylic acid anhydride in the above-described unsaturated ester compound, elevating the solution in pressure by means of a pressure pump, and introducing the solution directly into a vessel type reactor or a tubular reactor. Such a high-pressure polymerization process has been described namely in British Patent No. 2,091,745 as well as United States Patent No. 4,617,368 and United States Patent No. 4,644,044. The thus obtained ethylene copolymer has a melt index (as determined according to ASTM D-1238 from 1 to 500, and preferably from 3 to 200 g/10 min.

- Fillers to be used in the present invention, either organic or inorganic, are not particularly restricted, and preferably include inorganic fillers, such as calcium carbonate, talc, clay, silica, diatomaceous earth, alumina, zinc oxide, magnesium oxide, mica, calcium sulfite, calcium sulfate, calcium silicate, glass beads, glass fibers (inclusive of silane-treated glass fibers), asbestos, gypsum fibers, and the like ; and organic

fillers, such as various kinds of woodmeals, cellulose fibers, melamine powders, and the like. These fillers may be used either individually or, if desired, in combination thereof. In view of a favorable balance of impact resistance and heat resistance of the resulting filler-containing polypropylene resin composition, talc and/or mica are preferably used as fillers.

If desired, the filler to be used in the present invention can be subjected to surface treatment to ensure a good balance between rigidity and impact strength. Examples of the surface treating agent to be used are organosilane compounds including vinyltriethoxysilane, vinyltris( $\beta$ -methoxyethoxy)silane,  $\gamma$ -methacryloxypropyltrimethoxysilane,  $\gamma$ -aminopropyltriethoxysilane,  $\gamma$ -aminopropyltrimethoxysilane, N-( $\beta$ -aminoethyl)- $\gamma$ -aminopropylmethoxytrimethoxysilane,  $\gamma$ -glycidoxymethoxysilane, etc. Of these, N-( $\beta$ -aminoethyl)- $\gamma$ -aminopropylmethoxysilane and N-( $\beta$ -aminoethyl)- $\gamma$ -aminopropylmethoxytrimethoxysilane are preferred because of their high reactivity to produce a filler-containing polypropylene resin composition of highest level in balance between rigidity and impact strength.

The surface treating agent is usually used in an amount of from 0.1 to 2 part by weight, and preferably from 0.3 to 1 part by weight, per 100 parts by weight of the filler. If the amount of the surface treating agent is less than 0.1 part by weight, the bonding strength between the component (a) and the components (b) and (d) is insufficient so that the effect of the filler to improve rigidity cannot be produced and the impact strength becomes lower. If it exceeds the upper limit, the effect of the surface treating agent uneconomically comes to saturation. The surface treatment is preferably performed by the use of a ribbon mixer, a supermixer, etc.

The proportion of the component (a) in the composition of the present invention ranges from 27 to 87 % by weight. If it is less than 27 % by weight, heat resistance and rigidity as expected cannot be obtained.

The proportion of the component (c) in the composition ranges from 5 to 30 % by weight, and preferably from 10 to 20 % by weight.

The proportions of the component (d) in the composition is from 5 to 40 % by weight. If it is less than 5 % by weight, improved rigidity that is an essential requirement of the composition according to the present invention cannot be ensured. On the other hand, proportions exceeding 40 % by weight seriously reduce impact resistance of the composition, resulting in poor utility as materials for interior and exterior automobile parts. No further effects in improving thermal characteristics are produced by such an excessive amount of filler.

In the preparation of the filler-containing polypropylene composition in accordance with the present invention, the component (a) to (d) may be melt-kneaded all at once, but in a preferred embodiment, the kneading is carried out in two divided steps. That is, the components (a), (b) and (d) are melt-mixed to prepare (e) a mixture in the first step. The mixture (e) is then melt-kneaded with the component (c) to obtain the resin composition of the present invention. By this embodiment, a good balance of physical properties of molded products can be further ensured.

If desired, the filler-containing polypropylene resin composition of the present invention can further contain various styrene-based elastomers, such as a styrene-butadiene random copolymer, a styrene-butadiene block copolymer, and hydrogenated products thereof; various polyolefin elastomers, such as a non-crystalline ethylene-propylene copolymer and an ethylene-propylene-dicyclopentadiene terpolymer; and, in addition, various functional group-containing polyolefins, such as a glycidyl group-containing ethylene copolymer; as long as various characteristics possessed by the resin composition of the invention are not substantially impaired.

The resin composition of the present invention can furthermore contain appropriate amounts of various additives, such as antioxidants, weathering agents, antistatics, coloring agents, colorants, and the like.

For mixing the components, apparatus commonly employed for plastics or rubbers, such as a Banbury mixer, a single-screw extruder, a twin-screw extruder, and a roll mill, can be used. Considering that the resin composition in a molten state shows adhesion to metal surfaces due to its polarity, mixing is desirably carried out by means of an extruder. In particular, an extruder having two or more feed zones is economically beneficial because the first and second melt-kneading can be effected through one operation.

The resin composition obtained by the present invention can be molded into desired molded products by injection molding, extrusion molding, blow molding, etc., in a known manner.

The filler-containing polypropylene resin composition in accordance with the present invention possesses excellent finishing properties, such as adhesion, coating properties and printability, as well as excellent mechanical properties. In addition, the following advantages can be drawn from the resin composition of the present invention:

- The resin composition can produce injection molded products having a complicated shape and a small thickness under conventional molding conditions while substantially retaining flowability inherent to filler-containing polypropylene resins.

- the resin composition does not suffer from great reduction in mechanical characteristics, such as rigidity. In particular, the composition has markedly improved impact strength.  
 - weldlines (a defect of molded products caused by incomplete fusion of two or more flows of molten resins) are not so conspicuous as in the case of using the conventional filler-containing polypropylene resins.

Since the filler-containing polypropylene resin compositions according to the present invention bring about excellent effects as set forth above, they can be used in any field requiring finishing, such as adhesion, coating, and printing. Typical examples include exterior parts of automobiles, e.g., a bumper, a bumper corner, etc., interior parts of automobiles, e.g., an instrument board, a meter case, a glove compartment, a console box, a trim, a pillar-post, etc., motorcycle parts, e.g., a fender, a leg seal, etc., and appliance parts, e.g., a lid of a washing machine, a body of a vacuum cleaner, a stand of an electric fan, a frame of a ventilating fan, etc.

This invention will now be illustrated in greater details by way of the following examples, but it should be understood that they are not intended to limit the present invention. All proportions in these examples are given by weight unless otherwise indicated.

In these examples, an ethylene copolymer as component (c) was prepared as follows:

Ethylene containing a small amount of propane as a chain transfer agent and maleic anhydride were dissolved in an unsaturated ester compound, and the solution was continuously fed under pressure to a 1.2 l volume reaction vessel equipped with a stirrer. Copolymerization was effected at a temperature controlled to 180 °C under a pressure of 1800 kg/cm<sup>2</sup> at a rate of 1.5 to 3.2 kg/hr to obtain an ethylene copolymer.

Evaluations of physical properties of the resin compositions were made in accordance with the following test methods:

1) Surface free energy: specimens under test were prepared by molding the resin composition into a plate of 150 mm × 90 mm × 2 mm by the use of an in-line screw type injection molding machine of 69 g (manufactured by NISSEI JUSHI KOGYO K.K.).

2) Adhesive Strength: the same specimen as used in 1) above was coated with about 150 g/m<sup>2</sup> of an adhesive comprising 87 % by weight of G 250 (produced by CEMEDINE Co.Ltd.) and 3 % by weight of Desmodur (produced by BAYER AG) with an air-spray gun, followed by heating at 80 °C for 2 minutes. A skin material (PPX, produced by TORAY Industries Inc.) was heated at 110 °C for 2 minutes and, immediately thereafter, adhered to the adhesive-coated test plate under a planer pressure of 0.2 kg/cm<sup>2</sup>. After the laminate was allowed to stand at 23 °C and 65 % relative humidity for 7 days, peel test was conducted to measure a peel strength.

3) Initial Adhesion: The same specimen as used in 1) above was cleaned with 1,1,1-trichloroethane vapors (74 °C) for 30 minutes, followed by drying at room temperature. A urethane coat (Flexthane® #101, produced by NIPPON BEE CHEMICAL Co.Ltd.) was directly spray-coated on the plate and then baked in an oven at 120 °C for 30 minutes. The coating film was cross-hatched with a blade to make 2-millimeter squares (100 squares; 10 by 10). A 24 mm wide adhesive tape (Cellotape®, produced by NICHIBAN Co.Ltd.) was adhered thereon by pressing with fingers, and one end thereof was rapidly peeled off. The number of the squares remaining on the plate was counted to determine a percent remaining.

4) Izod notched Impact Strength: measured in accordance with standard ASTM D-256 at 23 °C.

5) Modulus in Flexure: measured in accordance with standard ASTM D-790 at 23 °C.

#### EXAMPLES 1 TO 5 COMPARATIVE EXAMPLES 1 TO 5

Ethylene copolymers (a) to (d) were prepared using maleic anhydride and an unsaturated ester compound as shown in Table 1 below.

TABLE 1

Sample No.	Comonomer Content (mol%)		Melt Index (190°C, g/10 min)
	Maleic Anhydride	Unsaturated Ester Compound	
(a)	1.22	butyl acrylate	5.34
(b)	1.10	2-ethylhexyl methacrylate	2.81
(c)	2.21	ethyl acrylate	7.52
(d)	1.95	methyl methacrylate	14.4

A polypropylene resin (Sumitomo Noblen® AW 584, a trade name for an ethylene/propylene block copolymer produced by Sumitomo Chemical Co., Ltd.; melt index: 8 g/10 min; ethylene content: 6 %; ethylene content in the propylene/ethylene copolymer unit 40%; intrinsic viscosity of the propylene/ethylene copolymer unit in a tetralin solution at 135°C: 8 dl/g), a modified ethylene/propylene copolymer containing 0.3 mol% of maleic anhydride (ethylene content: 10%; hereinafter referred to as A), each of the ethylene copolymers (a) to (d), a filler [talc having a mean particle size (corresponding to a 50% cumulative distribution by sedimentation-type particle size distribution measurement) of 3.0 µm or mica having a mean particle size of 3.5 µm] were blended at a compounding ratio shown in Table 2, and 0.2 part of Irganox® 1010 (produced by Ciba-Geigy AG), 0.05 part of calcium stearate, and 0.05 part of Ultranox® 626 (produced by Borg-Warner Corp.) per 100 parts of the total amount of the aforesaid components were also blended in a dry blender.

The dry blend was melt-kneaded in a vented extruder having a diameter of 65 mm (L/D = 28, manufactured by Ikegai Iron Works, Ltd.) at a die temperature of 220°C to obtain a resin composition in the form of pellets.

Each of the resulting resin compositions was evaluated for physical properties according to the above-described test methods, and the results obtained are shown in Table 3.

As is apparent from the Table, the molded products obtained from the filler-containing polypropylene resin compositions according to the present invention exhibit excellent performance properties, such as adhesion, coating properties, and the like, as compared with the comparative samples.

TABLE 2

Example No.	Polypropylene Resin		Ethylene Copolymer		Modified Polypropylene		Filler	
	Kind	Amount (wt%)	Kind	Amount (wt%)	Kind	Amount (wt%)	Kind	Amount (wt%)
Example 1	Sumitomo Noblen® AW 564	45	(a)	15	A	10	talc	30
" 2	"	45	(b)	15	A	10	"	30
" 3	"	45	(c)	15	A	10	"	30
" 4	"	45	(d)	15	A	10	"	30
" 5	"	45	(c)	15	A	10	mica	30
Comparative Example 1	"	70	-	-	-	-	talc	30
" 2	"	50	(a)	15	-	-	"	30
" 3	"	60	(b)	15	-	-	"	30
" 4	"	60	(c)	15	-	-	"	30
" 5	"	60	(d)	15	-	-	"	30

TABLE 3

Example No.	Wet Tensile Strength (dyne/cm)	Adhesive Strength (g/25 mm)	Initial Adhesion (%)	Izod Impact Strength (kg.cm/cm)
Example 1	33	4200	100	8
" 2	33	4000	100	8
" 3	34	4500	100	9
" 4	34	4400	100	8
" 5	34	4500	100	6
Comparative Example 1	29	1500	0	4
" 2	31	3400	70	8
" 3	31	3000	60	7
" 4	31	3500	90	8
" 5	31	3300	80	8

## EXAMPLES 8 TO 22 AND COMPARATIVE EXAMPLE 6

Sumitomo Noblen® AW584, a modified ethylene/propylene copolymer having 0.3 mol% of maleic anhydride (ethylene content: 10%; hereinafter referred to as PP-II), a modified ethylene/propylene copolymer having 0.05 mol% of maleic anhydride (ethylene content 10%; hereinafter referred to as PP-III), each of the ethylene copolymers (a) to (d) as prepared in Example 1, a filler (taic having a mean particle size of 3.0  $\mu\text{m}$  and/or mica having a particle size of 3.5  $\mu\text{m}$ ), a surface treating agent for the filler hereinafter described, and 0.02 part of Irganox 1010, 0.05 part of calcium stearate, and 0.05 part of Ultrinox® 626 per 100 parts of the total amount of the above-described components were blended in a dry blender. The resulting blend was melt-kneaded by means of the same extruder as used in Example 1 to obtain a filler-containing polypropylene resin composition.

The surface treating agent for the filler used was  $\gamma$ -aminopropyltriethoxysilane (TSL 8331), N-( $\beta$ -aminoethyl)- $\gamma$ -aminopropyltrimethoxysilane (TSL 8340), N-( $\beta$ -aminoethyl)- $\gamma$ -aminopropylmethyldimethoxysilane (TSL 8345),  $\gamma$ -glycidoxypolytrimethoxysilane (TSL 8350), or  $\gamma$ -methacryloxypropyltrimethoxysilane (TSL 8370), all produced by Toshiba Silicone K.K.



In Examples 6 and 7 and Comparative Example 6, all the above components were melt-kneaded at a compounding ratio shown in Table 4 through one step.

In each of Examples 8 to 22, the components shown in the upper row of Table 4 were first melt-kneaded at a compounding ratio indicated therein to once obtain a pellet mixture, and the resulting mixture was then melt-kneaded with the components shown in the lower row of Table 4 at a compounding ratio shown therein. Taking Example 14 for instance, Sumitomo Ncblen® AW 564, PP-II, and talc having been treated with TSL 8340 (1% based on talc) were first melt-kneaded at a weight ratio of 55/5/30 to prepare a pellet mixture. In other words, the ratio of these three components based on 100 parts by weight of the resulting mixture was 61.1/5.6/33.3. The resulting pellet mixture was then melt-kneaded with the ethylene copolymer (c) at a weight ratio of 90/10.

Each of the resulting resin compositions was evaluated for physical properties, and the results obtained are shown in Table 5.

It can be seen from Table 5 that a good balance of physical properties can be ensured by kneading the compounding components in two divided steps.

TABLE 4

Example No.	Polypropylene Resin		Ethylene Copolymer		Modified Polypropylene		Filler		Surface Treatment Agent	
	Kind	Amount (wt%)	Kind	Amount (wt%)	Kind	Amount (wt%)	Kind	Amount (wt%)	Kind	Amount (wt%)
Example 6	AW 564	55	(c)	10	PP-II	5	talc	30	-	-
" 7	"	55	(c)	10	"	5	"	30	-	TSL8340 1.0
" 8	"	55	-	-	"	5	"	30	-	" 0.03
" 9	"	55	-	-	"	5	"	30	-	" 2.5
" 10	"	55	(c)	10	"	5	talc	30	-	-
" 11	"	59	(c)	10	"	1	"	30	-	TSL8340 1.0
" 12	"	45	-	-	PP-III	15	"	30	-	" 1.0
" 13	"	40	-	-	PP-II	20	"	30	-	" 1.0
" 14	"	55	-	-	"	5	"	30	-	" 1.0
" 15	"	55	-	-	"	5	"	30	-	TSL8345 1.0
" 16	"	55	-	-	"	5	"	30	-	TSL8331 1.0

TABLE 4 (cont'd.)

Example No.	Polypropylene Resin		Ethylene Copolymer		Modified Polypropylene		Filler		Surface Treating Agent			
	Kind	Amount (wt%)	Kind	Amount (wt%)	Kind	Amount (wt%)	Kind	Amount (wt%)	Kind	Amount (wt%)		
Example 17	AW 564	55	- (c)	10	PP-II	5	talc	30	-	TSL8350	1.0	
" 18	"	55	- (c)	10	"	5	"	30	-	TSL8370	1.0	
" 19	"	50	- (c)	10	"	10	"	25	mica	5	TSL8340	1.0
" 20	"	50	- (a)	10	"	10	"	25	"	5	"	1.0
" 21	"	50	- (b)	10	"	10	"	25	"	5	"	1.0
" 22	"	50	- (d)	10	"	10	"	25	"	5	"	1.0
Comparative Example 6	"	70	(c)	-	-	-	talc	30	-	-	-	-

Note: \* Based on the filler or fillers.

Table 5

Example No.	Wet Tensile Strength (dyne/cm)	Adhesive Strength (g/25 mm)	Initial Adhesion (%)	Izod Impact Strength (kg·cm/cm)	Heat Deformation Temperature (°C)	Modulus in Flexure (kg/cm <sup>2</sup> )
Example 6	32	3600	100	7	101	11500
" 7	32	3600	100	7	108	13300
" 8	32	3600	100	7	108	13500
" 9	32	3600	100	10	129	18500
" 10	32	3600	100	7	108	13300
" 11	32	3400	100	7	108	13100
" 12	32	3400	100	7	108	13500
" 13	33	4400	100	10	130	18800
" 14	32	3600	100	8	127	17200
" 15	32	3600	100	8	127	17000
" 16	32	3700	100	8	123	16100
" 17	32	3600	100	8	109	14200
" 18	32	3500	100	7	112	14800
" 19	33	4700	100	8	129	21100
" 20	33	4500	100	8	129	20500
" 21	33	4500	100	8	132	22000

Table 5 (cont'd.)

Example No.	Wet Tensile Strength (dyne/cm)	Adhesive Strength (g/25 mm)	Initial Adhesion (%)	Izod Impact Strength (kg·cm/cm)	Heat Deformation Temperature (°C)	Modulus in Flexure (kg/cm <sup>2</sup> )
Example 22	34	4800	100	8	130	21500
Comparative Example 6	29	1500	0	4	137	12800

As described above, the filler-containing polypropylene resin compositions according to the present invention and molded products obtained therefrom have improved finishing properties, such as adhesion, coating properties, and printability, combined with impact resistance and heat resistance, as compared with the conventional compositions.

While the invention has been described in detail and with reference to specific embodiments thereof, it

will be apparent to one skilled in the art that various changes and modifications can be made therein without departing from the spirit and scope thereof.

## 5 Claims

1. A filler-containing polypropylene resin composition comprising:

(a) from 27 to 87 % by weight of a polypropylene resin,

(b) from 3 to 15 % by weight of a modified polypropylene resin containing from 0.1 to 3 mol % of an unsaturated dicarboxylic acid or anhydride repeating unit,

(c) from 5 to 30 % by weight of an ethylene copolymer comprising from 82 to 99.7 mol % of an ethylene repeating unit, from 0 to 15 mol % of an ester repeating unit selected from alkyl acrylates and alkyl methacrylates having from 1 to 8 carbon atoms in the alkyl moiety thereof, and from 0.3 to 3 mol % of an unsaturated dicarboxylic acid anhydride repeating unit, and

(d) from 5 to 40 % by weight of a filler.

2. A filler-containing polypropylene resin composition according to claim 1, characterized in that said unsaturated dicarboxylic acid or anhydride repeating unit in the component (b) is a maleic anhydride repeating unit.

3. A filler-containing polypropylene resin composition according to claim 1 or claim 2, characterized in that said unsaturated dicarboxylic acid anhydride repeating unit in the component (c) is a maleic anhydride repeating unit.

4. A filler-containing polypropylene resin composition according to one of claims 1 to 3, characterized in that said filler is an inorganic filler selected from calcium carbonate, talc, clay, silica, diatomaceous earth, alumina, zinc oxide, magnesium oxide, mica, calcium sulfite, calcium sulfate, calcium silicate, glass beads, glass fibers, asbestos and gypsum fibers.

5. A filler-containing polypropylene resin according to one of claims 1 to 4, characterized in that said filler is subjected to surface treatment by means of an aminosilane compound.

6. A filler-containing polypropylene resin composition according to claim 5, characterized in that said aminosilane compound is N-( $\beta$ -aminocetyl)- $\gamma$ -aminopropyltrimethoxysilane and/or N-( $\beta$ -aminoethyl)- $\gamma$ -aminopropylmethyltrimethoxysilane.

7. A filler-containing polypropylene resin composition according to any of claims 1 to 6, characterized in that the melt index of the ethylene copolymer (c) is from 1 to 500 g/10 min.

8. A filler-containing polypropylene resin composition according to any of claims 1 to 7, characterized in that the melt index of the polypropylene resin (a) is from 0.5 to 100 g/10 min.

9. A process for producing a filler-containing polypropylene resin composition according to claim 1 which comprises melt-mixing the components (a), (b) and (d) in a first step and melt-mixing the resulting mixture with the component (c).

10. A molded product obtained by molding a filler-containing polypropylene resin composition according to any of claims 1 to 8.



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# EUROPEAN SEARCH REPORT

Application Number

EP 87 40 2293

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
X	US-A-4 650 818 (M. OKA et al.) * Abstract; claim 1; column 2, lines 34-53 *	1-4,10	C 08 L 23/12 C 08 L 51/06 C 08 L 23/08 C 08 K 3/00 //
X,D	EP-A-0 213 720 (SUMITOMO) * Abstract; column 4, line 54 - column 5, line 10 *	1-8,10	(C 08 L 23/12 C 08 L 23/08 C 08 L 51:06 C 08 K 3:00 )
X	EP-A-0 184 321 (SUMITOMO) * Abstract; page 9, line 12 - page 10, line 22 *	1-8,10	
A	EP-A-0 058 980 (E.I. DU PONT DE NEMOURS) * Abstract *	1	
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			C 08 L C 08 K
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 05-07-1988	Examiner GOOVAERTS R.E.
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons A : technological background O : non-written disclosure P : intermediate document	